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Operation of a Sulfuric Acid Plant Using Blended Copper Smelter Gases

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16. ABSTRACT SOx (SO2 and SO3) emissions from copper reverberatory furnace off-gases can possibly be controlled to a high degree by blending the off-gases with those from roasters and converters and using the combined streams as a feed to a sulfuric acid plant. The Bor (Yugoslavia) copper smelter experimented briefly with the technique. Bor reported that when reverberatory gases were introduced into the total gas mix, absorption of SO3 in the acid plant apparently decreased, and a visible acid mist plume was observed. Bor attributed this poor acid plant operation to the high CO2 content of the reverberatory furnace gases, a conclusion contrary to other known situations where acid plants operate well with CO2-containing feed gas. The report gives results of an investigation of the effect of CO2 in the feed gases to a sulfuric acid plant on plant performance and emission levels. Available data on gas blending experience in Bor was compared with the experience of U.S. acid plant builders to determine if CO2 generally has an adverse effect or if other factors probably cause the acid plume formation experienced at Bor. The report indicates that the plume during gas stream blending at the Bor smelter is probably caused by an excessive load of sulfuric acid mist imposed upon the wet electrostatic precipitator that receives gases from the acid plant cooling system.

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OPERATION
OF A SULFURIC ACID PLANT
USING BLENDED COPPER SMELTER GASES

by

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ACKNOWLEDGMENTS

This report describes an investigation by the Research Triangle Institute, Research Triangle Park, N.C. of the operation of a contact sulfuric acid plant at a copper smelter being fed with blended exit gases from roasters, reverberatory furnaces, and converters. The investigation focuses on the problem of a visible plume from the acid plant stack.

The investigation was carried out under Contract No. 68-02-1325. Mr. R. V. Hendriks of the Environmental Protection Agency's (EPA) Industrial Environmental Research Laboratory, Research Triangle Park, served as Project Officer.

The investigation was conducted in the Process Engineering Department with Mr. Ben H. Carpenter as Project Leader. Several individuals contributed to specific aspects of the study. Mr. Raj Swarup interpreted much of the technical literature. Mr. J. P. Dempsey was engineer in residence at the Bor Copper Smelter, where the gas blending trials were made, during its expansion in 1970 and 1971. He provided an appraisal of the circumstances that prevailed during the blending of gas streams fed to the acid plants. Mr. Tim J. Browder, a specialist in the design and operation of sulfuric acid plants, appraised the information available on the Bor plants relative to the problem of plume formation and suggested design features necessary for elimination of plumes. Mr. J. H. Buddenberg, of the Collier Carbon and Chemical Corporation, permitted their sulfuric acid plant to be observed in operation. Mr. J. R. Donovan of Monsanto's Enviro-Chem Systems, Inc., provided technical information on mist control features of this plant. Performance data for the mist controls used at Collier were supplied by the Los Angeles Air Pollution Control District.

Mr. Norman Plaks, Chief, EPA's Metallurgical Processes Branch, Industrial Environmental Research Laboratory, provided additional guidance to the investigation. Messrs. Doug Bell, Charles Darvin, and Fred Porter of EPA's Office of Air Quality Planning and Standards, provided helpful suggestion.

SECTION 1

CONCLUSIONS

A high degree of control of SO_x emissions at copper smelters can be obtained by blending reverberatory furnace gases with gases from roasters and converters and using the combined stream as feed to a sulfuric acid plant. The Bor Copper Smelter in Bor, Yugoslavia, experimented with this technique for a short time and reported that visible plumes of acid mist were emitted from their acid plant stack. This was attributed to the carbon dioxide present in the reverb gases, which was presumed to decrease the absorption of SO_3 with the unabsorbed SO_3 emitted as a mist.

The results of this study indicate that the visible plume produced at the acid plant at Bor, Yugoslavia, when reverberatory furnace gases were added to its feed stream were caused by factors other than the presence of CO_2. The visible plume could most likely have resulted from additional sulfuric acid mist loads imposed upon the wet electrostatic precipitator (ESP) that receives cooled smelter gases from the acid plant cooling system. This would increase the concentration of mist in the gas leaving the ESP, and hence increase the quantity of mist carried through the dryers to the converter. Any of the mist disassociated to SO_3 and water in the converter would recombine as very fine mist as the gas cooled, and be carried through the SO_3 absorber to the stack.

Several factors could have increased the mist content of the blended gases when the reverberatory furnace gases were included:

- The reverberatory furnace was operated on pulverized coal, had manual controls and instrumentation, and substantial in-leakage of air. The resulting excess oxygen at high temperatures is conducive to the oxidation of nitrogen to NO_x, which could promote the formation of SO_3 in the exit gases. The reaction of SO_3 with water would yield acid mist.

- In the gas blending trials, the reverberatory furnace gas was fed to the mixing tower for the acid plant feed gases without treatment for removal of fine particulates. Any particulate matter not removed in the waste heat boiler was evidently carried some distance through the ducts, in which it may have promoted the formation of SO_3 from the SO_2 present, contributing to increased mist.
- The furnace gas was usually fed to the acid plant feed gas tower only when gas from an iron pyrites roaster was also available. Since this roaster was operated intermittently, the reverberatory furnace gas was flowing through its duct intermittently, with time for cooling, reaction, and condensation therein between successive runs. Mist formed in the duct while its flow was shut off could have been carried into the acid plant when gas flow was resumed.

Representative samples of the Bor reverberatory furnace gases showed the following composition ranges over an eight-hour period: SO_2, 0.4 to 3.1 percent by volume; CO_2, 1.4 to 12.4 percent by volume; O_2, 5 to 18.8 percent by volume. The estimated range of CO_2 concentration in the mixed gases fed to the sulfuric acid is 0.4 to 4.7 percent.

Sulfuric acid plants processing refinery alkylation acids, waste hydrogen sulfide and sulfur also process CO_2-containing feed gases frequently in greater concentration than that found in the Bor reverberatory gas. The danger exists that traces of impurities in these feed materials, not the CO_2, might cause a highly visible stack plume. Data was obtained on one of these plants which continues to operate without excessive SO_2 or H_2SO_4 mist emissions. The plant has the following units not present in the Bor plant: a Brink mist eliminator after the absorber, designed for an efficiency of 90 percent on 0.3 to 3 micron particles; an ammonia scrubber to reduce SO_2 emissions to below 300 ppm; and a Brink demister which reduces particulates to 20 mg per normal cubic meter. The system operates without a plume most of the time. Opacity of the stack gases exceeded the California standard (< 40%) only once during a

full day's compliance testing. For the rest of the test period, opacity readings were less than this standard; the haze was blue or white and not the black color for which the measurement method was designed. The high efficiency mist eliminators appear to be necessary to eliminate sulfuric acid mist.

SECTION 2

RECOMMENDATIONS

At this point, the study seems to have established the causes of plume formation in the acid plant during gas blending trials at Bor. The available operating data do not, however, provide sufficient information about the conditions of operation of the absorber which is a very important factor. Further study of the design of the Bor acid plant, to quantify the performance expected of the mist-control equipment is recommended.

It is recognized that the gas blending trials were not entirely pre-planned experiments; therefore, no systematic data collection could be expected. However, the analysis presented herein indicates that gas blending can proceed without a visible acid plant plume if the reverberatory furnace gases are produced with controlled burning, properly cleaned, and if the acid plant is equipped with high efficiency mist eliminators. Reverberatory furnace gases at Bor are now being cleaned with an ESP. The possibility of conducting a planned trial of gas blending involving planned sampling should be explored for its mutual possible advantages.

SECTION 3

INTRODUCTION

At domestic copper smelters, reverberatory furnaces present a difficult problem in control of sulfur dioxide (SO_2) emissions. Typically, SO_2 concentrations in the exit gases are too low for autogeneous conversion to sulfuric acid in a vanadium-catalyzed process. Neither is an alternative approach to control completely acceptable at this time. New source performance standards currently exempt these furnaces from control. Their total emissions, however, include dusts high in arsenic and other toxic substances. Reverberatory furnaces thus present an overall problem requiring the development of new technology.

The Environmental Protection Agency is investigating the possibility that a high degree of control of sulfur oxides (SO_2 and SO_3) emissions from copper reverberatory furnace off-gases can be achieved by blending these gases with those from roasters and converters and using the combined streams as a feed to a sulfuric acid plant. The Bor copper smelter in Bor, Yugoslavia, experimented with this technique for a short time. They reported, however, that when reverberatory gases were introduced into the total gas mix, the absorption of SO_3 in the acid plant was apparently decreased, and a visible acid mist plume was observed. Federal standards for emissions from U. S. acid plants exclude those attached to smelters, but smelter standards limit the opacity of acid plant stack gases to 20 percent. Thus, a visible plume would be poor operation in this Country. The Bor experts attributed this poor acid plant operation to the high carbon dioxide (CO_2) content of the reverberatory furnace gases. This conclusion is contrary to other known situations where acid plants operate well with CO_2 containing feed gas.

This study provides an investigation of the possible effect of CO_2 in the feed gases to a sulfuric acid plant on its performance and its emissions levels. Available data on the gas blending experience at Bor have been compiled and examined to determine the extent of knowledge concerning acid plant design, operating procedures, and gas stream conditions.

The information thus developed has been compared with the experience of American acid plant builders to determine whether CO_2 has generally an adverse effect or whether other factors are the likely cause of the acid plume formation experienced at Bor.

SECTION 4

GAS BLENDING AT BOR

4.1 THE SMELTER GAS-HANDLING SYSTEM

Figure 1 shows the gas-handling system at the Bor smelter at the time the first gas blending trials were made, in 1969. There were five Maguin multihearth roasters. Concentrates processes in the roasters contained about 0.8 percent arsenic. Roasting removed 45 to 50 percent of the sulfur. The resulting calcine was 17-18 percent sulfur. Gases left the roasters containing 8.8 to 10.6 percent SO_2 and 3.8 to 5.9 percent oxygen (O_2). The total volume averaged 50,000 Nm^3/hr. No external fuel was used. After dust removal and about 100 percent dilution, these gases typically contained about 5.7 percent SO_2 when combined with the converter gases.

Two of the plant's three "Pierce Smith" converters were operated at a time, and the gases were combined with the roaster gases after passing through electrostatic precipitators and blowers.

Gases just off the converters contained about 12 percent SO_2. The flow-rate from a single converter was about 33,000 Nm^3/hr at this point. After cleaning, with attendent dilutions, the gases averaged 2-6 percent SO_2.

The coal fired reverberatory furnace produced 50,000 kkg/yr of copper. Furnace gases were usually sent ot a stack but could be sent, via the duct which connected with the duct from a fluo-solids roaster, to a mixing tower for blending with gases from the roasters and converters. The composition of the reverberatory furnace gases at the time is not known, but the data for furnace operation in 1972 show a gas rate of 60,000 to 72,000 Nm^3/hr; with SO_2 content ranging from 0.4 to 3.1 percent by volume; CO_2, from 1.4 to 12.4; O_2 from 5 to 18.8.

Figure 1 shows a (Dorr-Oliver) fluo-solids roaster, which is mounted on top of the reverberatory furnace, with its exit gases passing through cyclones and an ESP, then through a duct to blending tower MT. This

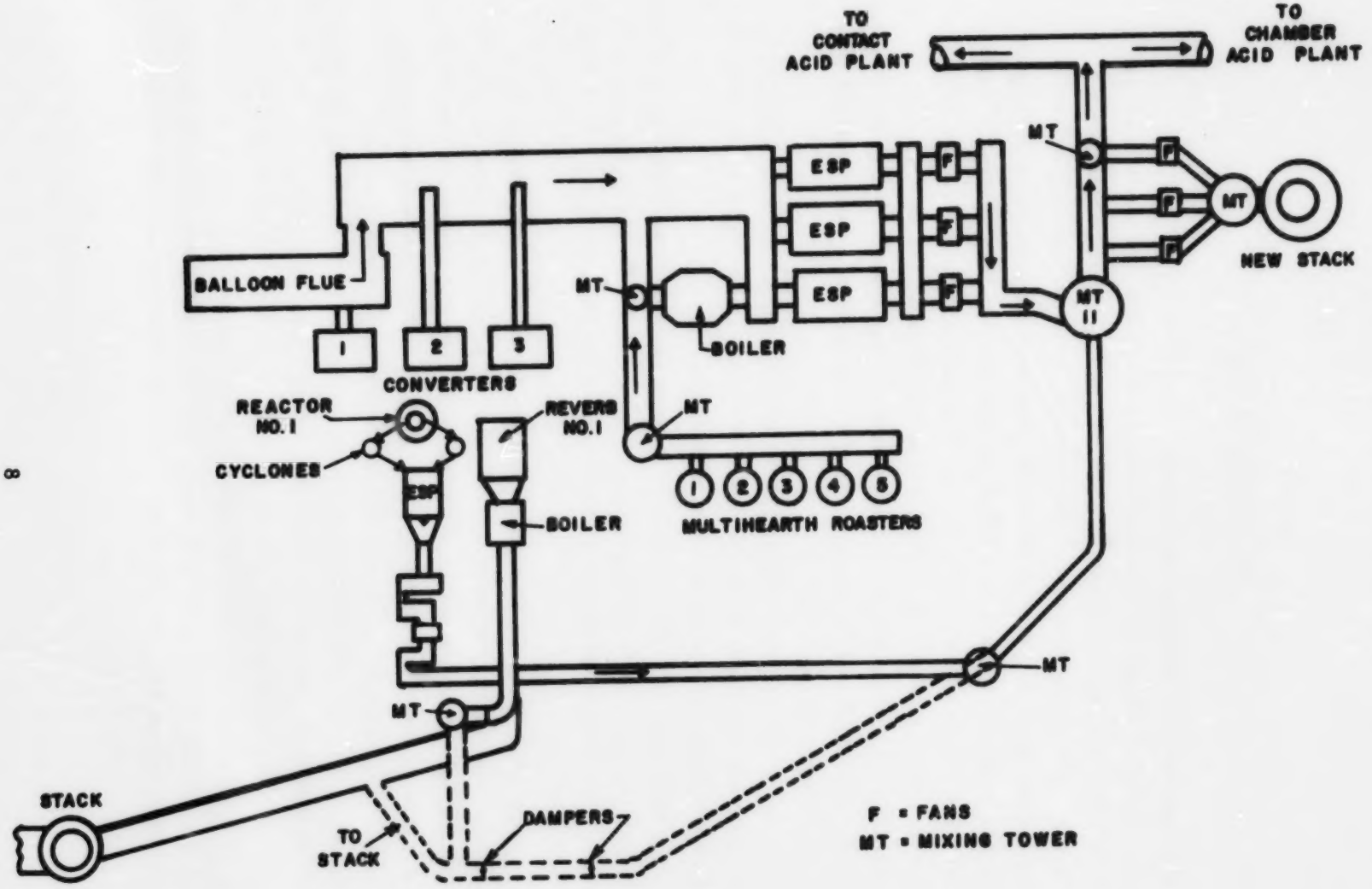

Figure 1. Gas handling system, Bor Plant.

roaster was operated half time, processing iron pyrites at a rate of 345 kkg/day. It produced some 48,000 Nm^3/hr* of gases containing 12-13 percent SO_2.

4.2 THE GAS-BLENDING TRIALS

In October 1969, the reverberatory furnace gases were mixed for a trial period with those of the multihearth roasters and converters, and fed to the acid plants, including the contact acid plant. During the trials, the mixture's SO_2 content was strengthened intermittently by addition of gases from the pyrites roaster. The gases were blended using the duct system shown in Figure 1, and were fed from mixing point MT_{11} to the sulfuric acid plant which at this time consisted of two chamber process units of Petersen design and one Chemiebau contact process unit. This study is concerned only with the contact unit. Gas rates there, after drying, ranged on a typical day from 81,000 to 83,500 Nm^3/hr. Since the total gas rate exceeded the capacity of the acid plants, excess gases were sent to a stack. By 1971, the Bor plant had added a second contact acid plant, similar to the first. Each of the contact acid plants had a sulfuric acid capacity of 430 kkg/day. A fluo-solids concentrate roaster was also in operation. In a second trial of gas blending, conducted in October, 1971, the feed to the acid plants included gases from two converters (3rd idle), the fluo-solids roaster and the multihearth roasters, the iron pyrites roaster, and one reverberatory furnace. Provision was made, as before, to heat the gases going into the contact plants, since the iron pyrites roaster was operated intermittently, and the SO_2 concentration of the feed gases was occasionally low when it was not operating. Feed rates to Contact Plant No. 1 ranged from 80,000 to 82,000 Nm^3/hr; those to Contact Plant No. 2, 88,000 to 90,000 Nm^3/hr.

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*The symbol N indicates standard conditions: 760 mmHg, and 273 K.

4.3 THE PERFORMANCE OF THE ACID PLANT

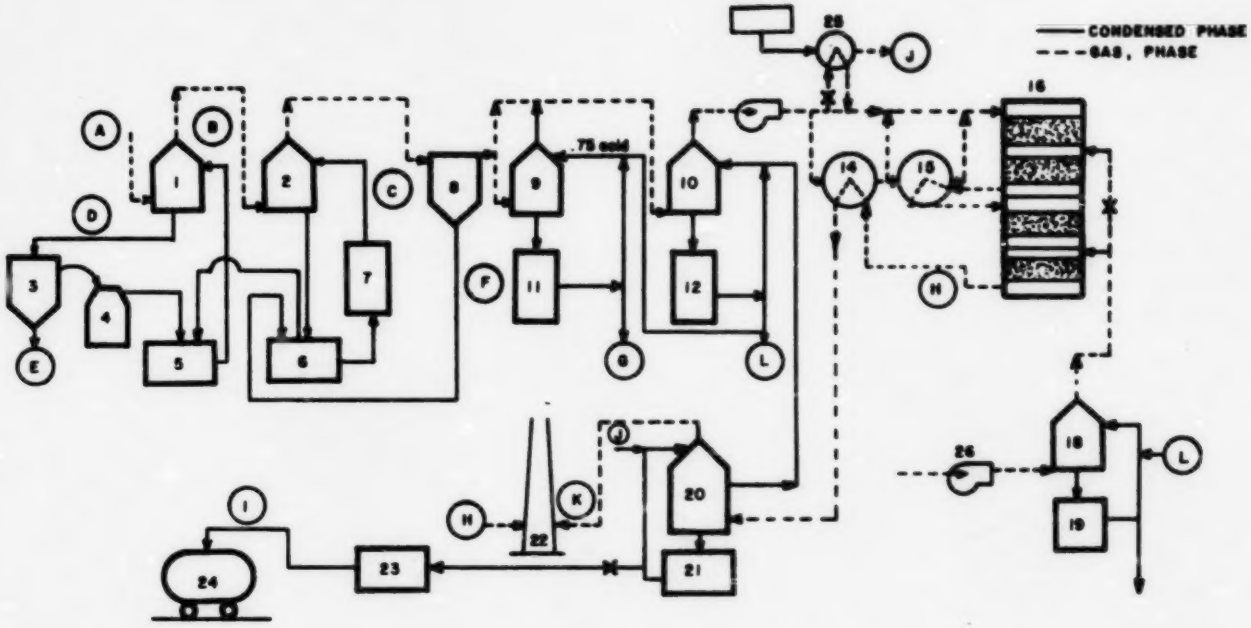
4.3.1 Operating Conditions

One contact plant was available for the trials in 1969; two, in 1971. Both plants were technically identical Chemibau units. The single absorption converters had four stages of vanadium oxide catalyst. The plant was comprised of four basic processes: gas purification, gas drying, oxidation of SO_2, and absorption of SO_3 (Figure 2). Operating data for two selected days for the first gas-blending trial and two days of the second trial are shown in Tables 1 to 8. The data in all these tables are given in the order: October 21, 1969; October 22, 1969; October 5, 1971: October 6, 1971, as indicated at the start of Table 1.

The gas purification process employed a steel gas cooling tower lined with acid resistant brick (1). Gases entering the plant with 12 to 17 percent oxygen (wet basis) were sprayed with 19 percent sulfuric acid, for cooling and removal of some dust. (The cooling operation is reported to produce sulfuric acid mist which absorbs catalyst-poisoning quantities of As_2O_3 and SeO_2 carried in the dusts). The gas pressures and temperatures in and out of the cooling tower are shown for every second hour of daily operation in Table 1. During the four days shown, gas temperatures of the exit gases ranged from 28 to 55°C. The average specific heat of gases entering the cooler is estimated to be 0.323 kcal/Nm^3.

Cooled gas passed to a Raschig-ring packed washing tower also constructed of brick-lined steel (2). Here it was sprayed with acid, part of which was recovered from the following ESP. Operating data for the washing tower are shown in Table 2. Exit gas temperatures ranged from 25 to 36°C.

Washed gas passed to a two-stage wet Electrostatic Precipitator (8). Operating data recorded for the ESP (Table 3) were gas pressure in and out, and data believed (pending receipt of further design information) to be concerned with control of the rectifier: the voltage on the high-voltage transformer (V), the current through power thyristors in series with



UNITS:
1. Gas Cooling Tower
2. Gas Washing Tower
3. Settling Tanks
4. Cooling Coils
5. Collecting Tank
6. Collecting Tank
7. Heat Exchanger
8. ESP
9. Pre-drying Tower
10. Drying Tower
11. Spray Coolers
12. Spray Cooler
13. Main Blower
14. Heat Exchanger
15. Heat Exchanger
16. Converter
17. Air Blower
18. Air Drying Tower
19. Spray Cooler
20. Absorption Tower
21. Spray Cooler
22. Stack
23. Storage Tanks
24. Cisterns
25. Oil-fired preheater (startup)
26. Air Blower

PROCESS STREAMS:
A. Smelter gases
B. Liquid feed to cooling tower
C. Liquid feed to washing tower
D. Effluent from Cooling tower
E. Slurry of solids and weak acid
F. Condensed mist from ESP
G. 75% H_2SO_4
H. Effluent from converter
I. 98% H_2SO_4
J. Make-up water
K. Absorption tower effluent
L. Transfer of acid from 12 to 18
M. Preheater exhaust to stack

Figure 2. Contact sulfuric acid plant, Bor, Yugoslavia

Table 1

Sulfuric Acid Plant Operating Data
Gas Purification Cooling Tower

Hour	Day	Gas Pressure, mmH$_2$O In	Gas Pressure, mmH$_2$O Out	Gas Temp. °C In	Gas Temp. °C Out	Acid Temp. °C In	Acid Temp. °C Out
6	10/21/69	110	215	260	52	28	49
	10/22/69	115	180	289	41	27	41
	10/ 5/71	0[1/]	-	198	40	35	48
	10/ 6/71	5	162	182	29	34	35
8		112	180	252	53	28	50
		40	90	182	38	29	41
		10	-	180	39	35	47
		15	72	200	30	35	35
10		110	185	240	55	29	50
		75	145	194	44	30	43
		5	-	175	41	31	47
		5	30	193	29	34	34
12		125	200	239	58	29	53
		70	150	205	45	30	47
		0	-	180	36	31	44
		15	57	200	29	35	36
14		85	175	220	47	28	46
		55	130	138	44	38	46
		4	-	198	40	37	46
		10	95	205	33	40	40
16		95	175	218	50	26	47
		65	140	208	45	37	46
		2	-	190	42	39	47
		25	110	210	30	33	33
18		100	230	235	50	30	51
		75	175	210	45	28	47
		20	-	178	41	39	46
		0	135	205	29	29	33
20		120	210	232	52	28	50
		80	170	206	48	27	44
		5	-	185	42	39	46
		10	160	202	28	28	31
22		90	175	240	50	28	48
		70	150	215	49	24	48
		10	-	190	39	32	44
		-	155	-	31	30	33
24		100	185	239	48	26	46
		65	195	222	53	30	50
		5	-	200	39	31	44
		-	155	-	31	32	35
2		130	185	218	45	28	47
		80	190	225	47	30	48
		15	-	200	40	31	46
		-	165	-	31	31	34
4		115	185	210	45	29	45
		80	170	220	48	28	40
		15	180	180	40	31	46
		-	75	-	31	31	34

[1/] Change in reading reflected in logs for 1971

Table 2

Sulfuric Acid Plant Operating Data
Gas Purification, Washing Tower

Hour	Gas Exit Temp. °C	Acid Temp. °C In	Acid Temp. °C Out
6	30	29	43
	29	22	38
	25	19	35
	27	20	34
8	35	23	43
	23	22	34
	25	19	33
	30	20	33
10	36	21	41
	23	25	33
	25	20	32
	30	24	34
12	36	21	43
	26	21	36
	26	21	33
	28	18	34
14	29	21	39
	26	23	37
	26	21	32
	31	18	38
16	27	19	37
	26	22	37
	27	22	33
	29	20	35
18	34	21	45
	31	25	39
	26	22	32
	20	16	36
20	33	20	45
	32	26	39
	26	22	32
	25	15	34
22	30	18	43
	32	30	43
	24	20	28
	32	21	35
24	29	17	42
	35	24	44
	26	22	29
	26	20	36
2	30	19	41
	32	20	41
	30	26	36
	29	18	35
4	34	20	40
	30	20	40
	25	20	35
	25	15	38

Table 3

Sulfuric Acid Plant Operating Data
Gas Purification
Wet Electrostatic Precipitator

Hour	Gas Pressure mm H_2O In	Out	First Stage Filters 1-4 V	A	mA	Second Stage Filters 1-2 V	A	mA	Filters 3-4 V	A	mA	Optical Visibility
6	275	365	260	30	95	260	0	40	240	5	20	good
	260	342	260	38	122	275	25	82	280	30	100	good
	215		292	32	110	310	34	110	275	32	100	good
	225		290	33	110	325	30	60	280	32	110	—
8	275	370	220	35	105	260	5	40	220	5	20	good
	110	205	240	32	100	230	15	20	250	10	55	good
	185		280	32	110	320	32	60	225	31	100	good
	260		280	26	130	325	29	60	290	36	125	—
10	265	370	260	30	95	260	10	40	220	10	50	good
	180	298	255	30	98	250	15	40	268	20	80	good
	187		290	33	110	325	32	60	280	31	100	good
	200		300	40	140	330	33	70	300	40	150	—
12	280	385	280	45	120	265	20	80	270	20	80	good
	188	307	250	30	100	220	25	80	270	30	100	good
	200		290	32	110	320	30	60	280	32	105	good
	265		300	40	145	330	33	65	295	42	150	—
14	245	340	278	45	130	278	25	100	272	30	100	good
	170	285	265	40	125	270	30	80	275	30	100	good
	210		287	33	110	330	30	60	280	32	105	good
	270		295	42	150	320	35	70	280	43	150	good
16	240	330	265	33	105	268	5	60	266	15	80	good
	195	310	258	30	98	280	30	100	280	30	100	good
	190		285	32	105	325	30	60	218	32	100	good
	218		200	40	140	320	34	65	283	42	142	good
18	340	430	266	33	105	240	15	50	265	15	58	good
	225	355	268	40	120	275	30	80	270	20	80	good
	320		290	34	110	325	30	60	290	30	100	good
	210		280	30	100	312	28	65	275	35	120	good
20	280	375	263	35	115	265	28	82	265	28	85	good
	225	355	262	26	40	260	15	60	270	10	60	fair
	205		282	32	105	320	29	60	275	31	100	good
	210		280	34	100	300	40	70	270	35	120	good
22	255	330	265	30	100	260	5	40	280	25	80	good
	210	325	270	35	120	280	30	100	280	30	100	good
	240		290	33	110	328	30	60	280	32	105	good
	260		290	35	120	305	25	55	265	30	100	good
24	255	330	285	45	140	270	35	105	265	30	100	good
	250	385	264	25	105	270	18	100	270	20	100	good
	250		285	33	110	325	30	60	280	32	110	good
	270		280	44	155	310	42	90	270	44	160	good
2	265	340	265	35	100	270	20	80	275	25	95	good
	235	355	270	35	100	280	35	100	280	30	100	good
	235		275	34	115	315	30	60	270	32	110	good
	300		295	42	150	215	41	90	275	42	150	good
4	253	335	260	24	100	268	18	80	274	20	100	good
	225	340	265	35	100	280	34	110	280	30	100	good
	235		280	33	110	320	29	60	275	32	110	good
	230		300	40	150	315	20	85	275	42	150	good

the high voltage transformer primary (A), and the value of high tension current supplying the electrical field (mA). Optical visibility at the ESP sight glass was classed as either good, bad, or fair, no bad readings were noted, but the observation was occasionally omitted.

The gas drying process employed a predrying tower and a drying tower. In the predrying tower (9), the gases were sprayed with strong sulfuric acid. Operating records for this tower (Table 4) indicate that the gases were washed with 82-76 percent acid at inlet temperatures from 30 to 36°C. The drying tower (10) to which the gases then passed was packed with Raschig rings. Here the gases were contacted with 93-97.8 percent acid at inlet temperatures of 28 to 39°C (Table 4).

Auxiliary air which was blown into sections of the converter as required for oxidation of SO_2 to SO_3, was also dried using a separate Rashig-ring packed tower (18). The data (Table 5) show a range of auxiliary air from 78-82 m^3/hr. This is about one percent of the SO_2-containing gas processed. Acid used ranged in concentration from 92 to 97.8 percent. Acid exit temperatures are given for three points, the relative positions of which are not yet defined. The range shown is 30-66°C, with the high values at exit 2.

From the drying tower, the gas was transferred by turbo-blowers into the converter system. A flow sheet for this system, derived from available information, is shown in Figure 3. Gases from the drying tower are heated by exchange with converter gases from the converter exit and from stage 2. Gas temperature in the converter is controlled by heat exchange and by auxiliary air blown into stages two and four. The auxiliary air also provides additional oxygen for conversion of SO_2 to SO_3. Operating data in Table 6 provides the performance of the first contacter during the 1969 gas blending trials, and the performance of the second converter in 1971.

Table 7 shows the gas pressure drops through the converter stages and the heat exchangers.

The SO_3 containing gases from the converter, after heat exchange (heat exchanger 1, Figure 3) were passed into the brick-lined steel absorption tower. The tower was packed with Raschig rings. Acid accumulated in

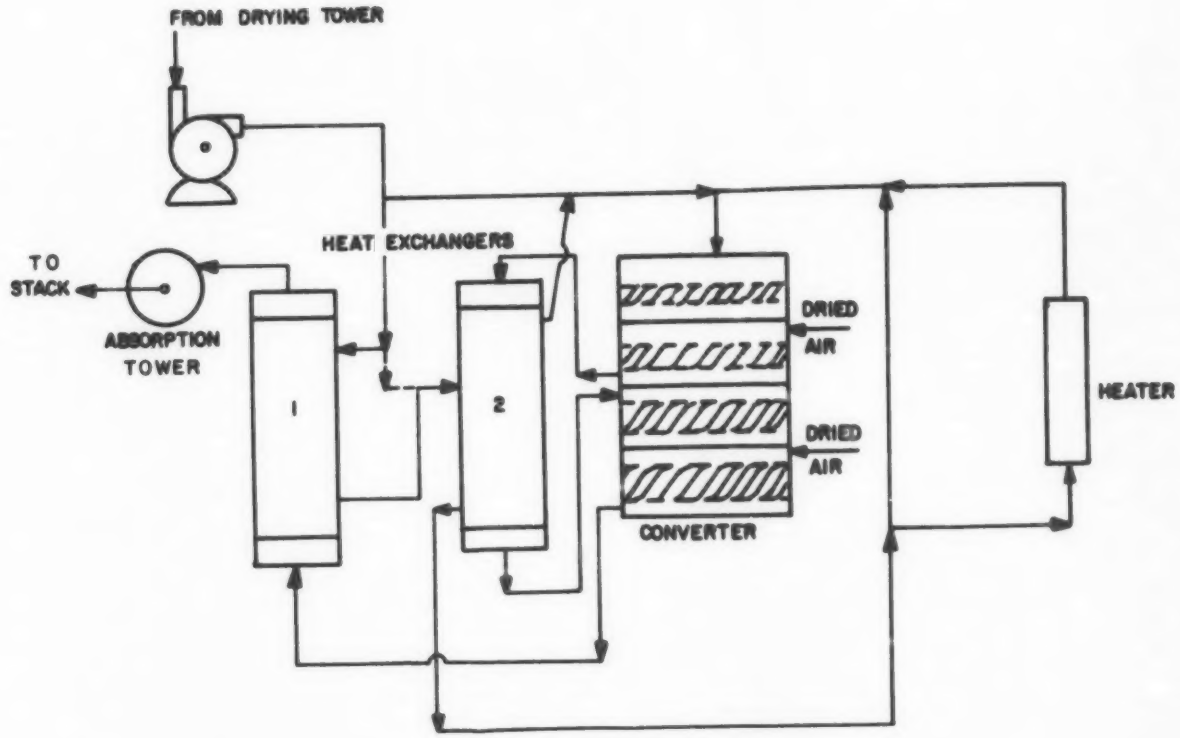


Figure 3. Gas flow sheet for sulfuric acid converter system, Bor, Yugoslavia.

Table 4

Sulfuric Acid Plant Operating Data
Gas Drying Towers

Hour	Acid: Predrying Tower Temp. °C In	Out	Density gm/cm³	Tank level	Drying Tower Temp. °C In	Out	m³/hr	Conc. %	level	Gas Pressure mm H_2O In	Out
6	30	35	1.660	610	32	44	-	97.53	810	345	460
	30	34	1.652	630	34	42	-	97.91	840	320	430
	30	45	1.680	560	28	36	-	93.00	1130	35	200
	33	-	1.5456	740	30	46	-	93.00	1280	25	205
8	30	35	1.660	700	34	50	-	97.18	805	350	465
	30	33	1.660	690	30	42	-	97.67	920	188	270
	30	45	1.680	700	32	46	-	93.00	1320	20	210
	35	-	1.5368	740	32	48	-	93.00	1320	25	205
10	29	35	1.655	600	35	50	-	97.10	840	355	465
	33	36	1.658	700	31	44	-	97.59	810	275	388
	30	45	1.660	710	33	46	-	94.90	1420	20	215
	34	-	1.5866	840	30	44	-	93.00	1270	23	208
12	30	36	1.660	680	35	50	-	97.19	850	365	485
	31	35	1.660	700	32	42	-	97.60	800	286	390
	30	45	1.660	760	32	45	-	95.95	1240	23	208
	30	-	1.4876	1000	31	44	-	93.00	1070	23	210
14	30	34	1.660	680	34	44	-	97.50	770	325	435
	30	35	1.660	660	34	44	-	97.60	790	265	370
	30	44	1.660	700	33	48	-	96.02	960	25	208
	33	-	1.5446	930	33	48	-	93.00	1260	23	210
16	33	33	1.660	630	32	42	-	97.77	880	300	410
	30	35	1.660	670	34	45	-	97.69	800	230	395
	31	46	1.652	830	35	50	-	96.20	1370	21	208
	32	-		760	33	46	-	93.00	1250	21	207
18	32	40	1.662	690	35	52	-	97.50	860	405	530
	30	37	1.660	650	35	48	-	97.50	840	325	450
	32	47	1.650	900	37	52	-	95.34	1070	23	205
	30	48	1.660	1000	30	70	-	93.00	1270	22	210
20	30	38	1.660	600	33	49	-	97.37	800	360	465
	31	36	1.660	600	35	50	-	97.36	800	325	455
	33	50	1.656	820	38	55	-	95.10	1070	22	205
	36	44	1.664	900	29	38	-	93.00	1120	21	207
22	34	40	1.662	690	34	46	-	97.36	820	315	420
	30	37	1.654	680	33	48	-	97.80	740	319	425
	30	45	1.652	870	35	50	-	95.39	1190	28	205
	32	52	1.644	810	30	43	-	93.00	1200	27	210
24	29	34	1.662	580	34	46	-	97.37	830	310	415
	35	40	1.660	640	34	48	-	97.16	790	350	475
	33	48	1.655	720	35	50	-	95.81	1160	30	200
	35	61	1.650	800	39	52	-	93.00	1280	23	200
2	29	32	1.660	560	32	40	-	97.65	650	320	430
	32	39	1.666	660	33	46	-	97.39	780	330	455
	32	54	1.650	680	32	56	-	93.00	1080	25	205
	35	55	1.652	700	33	46	-	93.00	1180	27	210
4	31	36	1.660	680	31	48	-	97.79	750	320	420
	31	35	1.650	700	32	43	-	97.45	760	320	435
	30	50	1.658	780	30	43	-	93.00	1240	27	210
	33	52	1.650	600	31	42	-	93.00	1130	22	205

Table 5

Sulfuric Acid Plant Operating Data
Auxiliary Air Drying Tower

Hour	Acid Temp. °C Exit 1	Exit 2	Exit 3	Feed Rate m^3/hr	Conc. %
6	30	32	30	82	97.54
	31	35	34	82	97.75
	36	55	31	-	92.56
	34	54	33	-	96.42
8	29	32	32	80	97.26
	30	34	32	80	97.68
	28	49	35	-	94.90
	38	58	35	-	96.45
10	32	35	35	80	97.15
	31	33	32	80	97.67
	29	48	37	-	96.29
	38	56	33	-	96.15
12	34	35	35	80	97.20
	30	30	30	80	97.65
	28	48	36	-	96.86
	38	58	34	-	95.72
14	30	34	34	80	97.42
	30	33	32	80	97.61
	28	40	37	-	96.94
	41	66	35	-	96.52
16	31	34	33	81	97.15
	30	35	34	78	97.67
	29	41	38	-	97.07
	37	56	36	-	96.12
18	32	35	35	80	97.52
	30	34	35	80	97.59
	31	43	40	-	96.38
	35	52	30	-	97.18
20	30	34	33	79	97.41
	30	36	35	79	97.36
	32	44	42	-	96.18
	30	47	30	-	94.70
22	28	34	32	78	97.35
	29	34	32	80	97.05
	29	41	39	-	96.71
	34	58	31	-	95.69
24	34	38	35	79	97.56
	30	35	33	80	97.16
	31	41	37	-	96.68
	41	62	40	-	95.50
2	32	30	30	80	97.65
	30	37	30	78	97.28
	31	43	39	-	95.17
	38	56	38	-	95.12
4	30	35	30	80	97.74
	29	34	30	80	97.45
	32	48	33	-	96.12
	37	56	33	-	95.10

Table 6

Sulfuric Acid Plant Operating Data
Converters

Hour	Gas Rate 1000 Nm³/hr	Gas SO_2 %	Added Air[1] 1000 Nm³/hr S1/S2[2]	S3/S4	TEMPERATURE, °C Outer Heat Exchanger In	Out	Exit B'tws Exc's.	Stage 1 In	Out	Stage 2 In	Out	Stage 3 In	Out	Stage 4 In	Out	Exit Outer Heat Exc.
6	82	4.0	8.5	5.	71	365	505	468	572	532	543	482	470	458	458	195
	82	3.3	3.0	1.5	70	339	479	441	541	527	526[1]	435	445	435	435	178
	90[1]	2.8	-[3]	-	75	335	422	440	520	520	520	430	438	438	430	160
	90	3.8	-	-	68	350	448	455	560	542	542	457	460	450	445	182
8	82	3.9	9.	5.	70	350	490	452	560	522	525	450	455	438	438	185
	70	0.7	0.	0.	61	322	418	427	473	472	490	422	447	428	429	157
	92	3.2	-	-	75	322	412	421	505	505	506	419	424	421	423	160
	90	4.0	-	-	80	354	440	460	590	580	570	460	473	457	495	185
10	82.7	4.1	7.2	4.9	71	340	483	449	556	520	526	445	450	435	435	182
	81.0	4.0	0.	0.	71	311	445	412	508	496	481[1]	409	412	412	417	155
	92.5	4.9	-	-	78	322	420	431	538	530	525	422	440	440	435	160
	90	4.4	-	-	75	360	440	455	560	530	530	455	458	450	450	185
12	82	5.4	9.	4.6	75	340	480	450	565	525	530	445	450	438	438	185
	80	2.9	0.	0.	67	310	465	429	519	511	518	422	425	425	425	155
	91	5.3	-	-	75	315	423	434	535	532	530	428	440	441	440	162
	90	3.4	-	-	75	345	440	445	547	545	542	442	460	460	447	162
14	83.5	2.5(P)	1.	4.9	72	340	480	448	549	523	528	438	449	431	431	171
	80	2.4	0.	0.	68	328	468	432	512	508	518	425	428	428	428	155
	90	3.9	-	-	72	347	446	455	510	569	560	447	460	460	452	167
	90	6.5	-	-	75	373	452	467	598	572	572	470	470	467	453	198
16	82	4.9(P)	2.7	4.7	67	332	480	444	550	524	530	434	444	428	432	170
	80	5.2	9.2	1.1	70	330	482	442	560	520	532	438	440	432	435	170
	81	3.9	-	-	75	344	438	450	572	577	597	472	452	452	450	167
	90	8.3	-	-	73	387	447	463	593	547	548	477	488	488	478	210

Table 6 (Cont.)

Hour	Gas Rate 1000 Nm³/hr	Gas SO_2 %	Added Air[1] 1000 Nm³/hr S1/S2[2]	Added Air[1] 1000 Nm³/hr S3/S4	Temperature, °C: Outer Heat Exchanger In	Outer Heat Exchanger Out	Exit B'tvn Exc's.	Stage 1 In	Stage 1 Out	Stage 2 In	Stage 2 Out	Stage 3 In	Stage 3 Out	Stage 4 In	Stage 4 Out	Exit Outer Heat Exc.
18	88	6.2(P)	10.	5.0	71	360	498	457	586	545	552	445	462	440	442	191
	86	4.2	3.9	3.8	70	332	482	448	552	522	530	440	448	432	435	175
	90	4.2	-	-	78	373	442	448	560	560	558	448	460	760	755	170
	90	3.7	-	-	70	372	429	422	540	527	525	458	462	460	460	192
20	83	4.2(P)	9.6	5.0	67	358	480	448	560	516	525	436	449	428	432	190
	88	4.1(P)	8.0	5.5	70	346	488	452	562	520	530	450	458	440	435	180
	91	4.8	-	-	80	353	452	463	572	572	567	456	471	471	465	172
	90	5.4	-	-	70	363	445	460	580	545	548	460	470	470	460	182
22	82	3.2(P)	4.8	4.8	68	348	482	430	550	525	527	440	448	428	430	185
	85	4.8(P)	9.2	5.0	70	350	491	457	569	522	532	450	458	438	439	185
	90	6.0	-	-	79	358	450	461	580	580	563	458	472	472	462	170
	90	5.2	-	-	72	362	445	469	570	545	545	466	472	472	468	192
24	81	3.6(P)	7.5	4.5	69	341	438	450	560	525	530	440	450	430	432	180
	85	4.5(P)	10.2	5.0	70	352	492	455	571	531	540	445	459	436	440	190
	90	3.2	-	-	82	362	462	470	588	586	579	465	484	484	473	175
	88	5.2	-	-	78	385	441	460	582	530	540	470	482	482	475	215
2	81	3.9	4.8	3.9	68	342	482	447	560	522	530	433	448	430	432	178
	85	5.0	3.5	5.0	70	360	490	450	570	525	531	444	450	439	443	190
	88	3.3	-	-	85	375	459	470	572	555	555	465	478	458	452	198
	90	8.0	-	-	78	380	445	460	595	555	558	422	435	424	465	210
4	82	2.2	0.	5.8	65	340	479	441	525	521	521	430	444	428	430	175
	85	3.4	3.7	7.0	68	355	487	450	550	525	522	440	458	430	440	188
	91	4.9	-	-	75	350	438	450	562	538	532	445	458	458	439	178
	90	4.9	-	-	79	332	448	465	590	560	560	428	490	479	472	215

[1] First . days shown are for the first contact acid plant; the last two, for the second (newer) acid plant.

(P) Indicates that the feed gas contained iron pyrites roaster gases.

[2] S1/S2 denotes air added between the first and second catalyst beds of the converter. S3/S4 denotes air added between the third and fourth bed.

[3] No data recorded.

Table 7

Sulfuric Plant Operating Data
Converters

Hour	Outermost Heat Exc. Points 1	2	11	Pressure, mm H_2O Stage 1 In	Out	Stage 2 In	Out	Stage 3 In	Out	Stage 4 In	Out
6	1900	1420	80	1260	1155	1080	975	780	645	613	428
	1610	1168	60	1041	933	868	776	625	522	489	278
	2400	-	-	1820	1440	1390	1260	1050	846	790	432
	2700	-	-	2270	1710	1610	1440	1220	1000	960	620
8	1880	1394	79	1244	1140	1066	962	786	660	613	423
	1250	883	40	770	690	638	572	450	377	340	220
	2420	-	-	1610	1440	1382	1256	1034	871	790	505
	2690	-	-	1910	1740	1080	1540	1290	1060	1000	945
10	1820	1360	73	1208	1108	1039	928	746	635	586	408
	1500	1078	55	944	862	776	692	494	438	419	287
	2430	-	-	1630	1460	1382	1254	1020	832	780	495
	2730	-	-	1820	1760	1690	1660	1300	1040	1010	660
12	1850	1380	75	1214	1108	1040	922	746	632	583	408
	1590	1112	63	992	888	820	730	584	780	448	300
	2430	-	-	1630	1444	1384	1250	1022	830	774	495
	2500	-	-	1680	1570	1420	1300	1170	890	810	510
14	1790	1270	70	1122	1012	950	860	702	600	544	375
	1580	1130	60	990	884	820	734	672	485	440	303
	2410	-	-	1630	1456	1396	1269	1090	838	792	499
	2700	-	-	1910	1736	1673	1530	1280	1040	1005	640
16	1790	1280	68	1169	1070	912	818	752	624	525	363
	1780	1364	69	1100	994	910	822	710	652	505	348
	2400	-	-	1624	1460	1390	1260	1034	837	794	495
	2800	-	-	2020	1844	1790	1627	1374	1077	1025	960
18	2030	1570	82	1374	1260	1170	1064	860	735	684	475
	1800	1373	70	1200	1095	1002	910	730	620	570	397
	2400	-	-	1720	1390	1330	1200	990	800	750	470
	2600	-	-	1790	1624	1556	1424	1190	985	819	596
20	1850	1398	79	1250	1152	1080	962	780	668	517	398
	2020	1522	52	1380	1254	1152	1050	844	718	665	465
	2400	-	-	1620	1460	1390	1260	1030	815	750	488
	2700	-	-	1875	1700	1630	1480	1230	1000	945	600
22	1750	1256	70	1104	1000	934	832	672	563	515	352
	1990	1486	80	1306	1202	1118	1010	816	692	643	450
	2420	-	-	1630	1450	1380	1260	1030	810	775	490
	2700	-	-	1880	1720	1640	1500	1230	1015	950	605
24	1780	1306	67	1076	1004	996	906	730	618	568	324
	1970	1478	75	1310	1204	1194	1000	808	682	630	431
	2480	-	-	1660	1500	1410	1275	1040	825	780	490
	2680	-	-	1910	1760	1690	1550	1300	1045	1000	645
2	1710	1252	65	1110	1008	992	942	679	576	528	353
	1930	1470	85	1310	1200	1120	1020	830	700	645	440
	2600	-	-	1870	1630	1610	1490	1250	1020	985	640
	2760	-	-	2005	1820	1760	1610	1350	1115	1045	695
4	1650	1184	60	1048	948	880	708	632	533	490	340
	1890	1394	65	1294	1132	1050	842	773	650	616	428
	2600	-	-	1770	1600	1530	1410	1150	950	870	560
	2780	-	-	2005	1835	1760	1620	1350	1115	1045	680

the reservoir at the tower base at 98-99 percent concentration after being sprayed into the tower at about 500 m^3/hr (Table 8). The desorbed gases left the absorber at 60 to 83°C. The circulated acid was cooled by passing it through trough coolers sprayed with water. The gases were passed through a "drip separator" (not fully described), and then to a stack.

4.3.2 Results

The SO_2 content of the exit gases was estimated at 1500 ppm. When reverberatory furnace gases were added to the feed gas for treatment in the acid plant, the exit gases developed a plume of acid mist. This disappeared when these gases were taken out of the feed stream. The formation of the plume was attributed by the Yugoslav operators to the CO_2 content of the reverb gases. This was presumed to have decreased the absorption of SO_3. Unabsorbed SO_3 was then emitted to form a sulfuric acid mist.

Table 8

Sulfuric Acid Plant Operating Data
Absorber

Hour	Gas		Acid			
	Exit Temp. °C	mm H_2O	Temp. °C In	Temp. °C Out	m^3/hr	Conc. %
6	69	16	60	78	500	98.54
	66	8	60	74	490	98.49
	75	-	60	78	-	98.35
	80	-	64	84	-	98.48
8	65	15	60	74	510	98.60
	62	4	60	68	510	98.80
	77	-	63	78	-	97.83
	83	-	68	92	-	98.48
10	68	16	60	73	500	98.56
	68	7	68	70	500	98.50
	65	-	56	77	-	98.20
	83	-	68	90	-	98.42
12	72	14	62	78	500	98.59
	63	10	60	68	515	98.52
	68	-	52	73	-	98.98
	70	-	64	80	-	97.90
14	61	15	59	72	505	98.53
	62	10	58	68	510	98.58
	78	-	62	84	-	99.00
	80	-	70	94	-	98.50
16	64	16	60	76	510	98.64
	71	11	62	78	505	98.58
	76	-	62	80	-	98.68
	80	-	64	90	-	98.48
18	70	21	62	80	500	98.65
	65	15	60	74	500	98.56
	78	-	68	85	-	98.25
	70	-	60	82	-	98.61
20	64	16	60	76	520	98.51
	62	17	60	78	515	98.54
	81	-	67	85	-	98.12
	70	-	58	80	-	98.27
22	61	12	58	71	500	98.54
	68	16	62	78	510	98.83
	79	-	63	83	-	98.67
	79	-	61	83	-	98.54
24	70	13	62	76	490	98.60
	70	12	62	76	500	98.60
	80	-	62	83	-	98.63
	80	-	64	88	-	98.26
2	62	14	60	72	495	98.62
	65	15	59	74	540	98.58
	79	-	63	85	-	98.41
	80	-	62	90	-	98.44
4	60	13	58	70	500	98.57
	60	13	58	72	520	98.54
	72	-	59	80	-	98.48
	78	-	62	90	-	98.41

SECTION 5

EXAMINATION OF THE FACTORS RELATING TO THE PLUME FORMATION

5.1 SURVEY OF EXPERIENCE WITH CO_2-CONTAINING FEED GAS

Several sulfuric acid plant designers and builders have indicated that some of their plants handle CO_2-containing feed gases without a plume in the exit gases. While very high CO_2 concentrations were felt to be adverse to over all operation, there was no valid scientific or experimental support for this thinking. The estimated range of CO_2 concentration in the mixed gases fed to the acid plant at Bor is 0.4 to 4.7 percent. This is lower than the 5 to 6 percent CO_2 in the feed to sludge burning plants.

Many firms had experienced plume problems or operation upsets due to 1) nitrogen oxide in the feed gases, 2) CO from incompletely burned organic matter, 3) excessive moisture in the gases entering the converter, 4) insufficient rate of acid circulation and lack of uniform distribution in the SO_3 absorption tower, 5) unclean packing in the SO_3 absorption tower, 6) improper concentration and temperature of the absorbing acid, and 7) shock cooling of the gases leaving the converter below their dew point.

Table 9 summarizes the experiences and opinions of the several firms with whom the Bor gas-blending trial was discussed.

5.2 APPRAISAL OF CONDITIONS PREVAILING DURING TRIAL RUNS*

The conditions prevailing at the Bor plant during the two tests of gas blending offered several opportunities for generation of a plume: an intermittently used underground flue; excess combustion air; and excess particulate matter in the uncleaned reverberatory furnace gases.

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*Based on personal observations of Mr. J.P. Dempsey who was resident engineer during the Bor plant expansion in 1970 and 1971, and on analysis of the information by Mr. Tim J. Browder, specialist in sulfuric acid plant design.

Table 9

Experiences of Acid Plant Designers and Builders

Firm	
M. W. Kellogg	Sludge burning plants with 5-6% CO_2 in the feed gas have had no plume problems. NO_x in converter exit gas seems to promote formation of H_2SO_4 mist from SO_3 and H_2O before the gas reaches the absorber. Low flame temperatures suppress the plume, possibly because they supress NO_x.
Ralph M. Parsons	CO_2 has not been known to cause a plume. One satisfactory elemental sulfur conversion plant feeds a gas that tests 5-8% CO_2 and 0.9% O_2 at the stack.
Stauffer Chemicals Co.	CO_2 cannot cause a plume. Their alkylation acid plants process feed gases with greater than 4% CO_2 without plume problems.
Smelter Control Research Assoc.	The cause of plume formation is most likely H_2SO_4 mist.
Chemical Process Plants Division of Chemico	CO_2 has had no effect. Brink mist eliminators are normally used to remove any mists that might be present from whatever cause, and that might otherwise form a plume.
Fluor, Utah	CO_2 cannot cause a plume. NO_x probably promotes the formation of H_2SO_4 mists in the presence of moisture, and the mist causes the plume.
Leonard Construction Co.	CO_2 has not been a problem.
Enviro-Chem	CO_2 has not been a problem. Absorber acid concentration, temperature, and inadequate distribution can contribute to plume formation.
Picatinny Arsenal	Wire mesh types of mist eliminators do not control the plume from TNT plant acid recovery systems.

At the time of the 1969 tests, a flue conveyed gas from the No. 1 Reverberatory Furnace underground to the old stack. There was a connection to the flue from the pyrites roaster just prior to the point where the reverberatory flue went underground. Accordingly, the furnace gas was mixed with the pyrites roaster gas downstream from the roaster ESP. This means that the furnace exit gas still contained all particulate matter not removed in the waste-heat boiler. These particulates could have promoted the conversion of SO_2 in the gas mix to SO_3, which would then form excessive mists as the gas traveled on through the duct system.

The reverberatory furnace was a fairly old furnace, built in 1958-1959. It was fired with pulverized coal in 1969 and 1971, and had minimal process controls and instrumentation. The side wall brick had been replaced many times. In 1970 and 1971, these side walls looked like a sieve, and substantial air inleakage was unavoidable. Excess air during combustion in the furnace would have caused oxidation of nitrogen to NO_x which would catalyze the formation of SO_3 in the exit gases. The cooling and drying systems of the acid plant would not be expected to remove fine mist particles (< 3 microns) formed from SO_3 and water. These would then overload the ESP sufficiently to allow excessive mist to reach the converter, dissociate there, and reform in the absorber, overload the "drip separator", and exit to generate a plume.

The intermittant operation of the pyrites roaster would have provided an opportunity for acid mist to accumulate in that part of the duct system which was thus intermittently idle. This mist would have been swept into the acid plant when the roaster operation was resumed.

The cause of the plume is judged to have been the use of excessive air in burning the coal in the reverberatory furnace, leading to formation of NO_x which, with excessive particulate dust passed into the gas handling system. Here excessive sulfuric acid mists were formed which passed through the acid plant to the atmosphere.

5.3 DESIGN FEATURES THAT CONTROL PLUMES

Figure 4 shows the gas flow through an acid plant operated by the Collier Carbon and Chemical Corporation to produce sulfuric acid from refinery hydrogen sulfide, alkylation acid, and sulfur. The system feeds gases produced by burning the three sulfur bearing materials, and the feed is usually a composite form of all three. This plant was observed to operate without any plume formation. Its performance had been confirmed by tests carried out by the Los Angeles Air Pollution Control District. While limited data are available on the composition of the feed gases to this plant, the feed gas may be presumed to contain CO_2 at a somewhat higher concentration than the reverberatory furnace gases at Bor. Resen (37) gives the following composition of gases from a comparable sludge acid furnace: 30.7 percent H_2O, 8.6 percent SO_2, 9.7 percent CO_2, 1.5 percent O_2, 49.4 percent N_2. The Collier plant controls the excess O_2 in the burner exit gases at 3 percent, which would reduce their SO_2 and CO_2 concentrations somewhat below those given for this sludge acid furnace described by Resen. The gas feed rate to the stack at the Collier plant was about 35,000 Nm^3/hr. (The converter feed gas showed 10 to 11.2 percent SO_2).

In the humidifying tower, the gas is washed with a 0 to 5 percent H_2SO_4 spray. It is then cooled in "Carbate" coolers, to 27-30°C, and some water and acid condense. The cooled gas is treated in an ESP which was fabricated using 15 cm diameter lead tubes. This unit has an estimated particle elimination efficiency of 95 - 99 percent (weight basis).

After further cooling, the gas is dried with 96 -97 percent H_2SO_4. A 98 percent acid would be too high; acid and SO_3 vapor pressures would be high enough to add mist to the gases.*

The plant has a gas-fired startup pre-heater which can be bypassed. Heat exchangers can also be bypassed but usually adjust to changes in flow by the changes in heat transfer coefficient thereby induced.

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*Discussion with Mr. J. R. Donovan of Monsanto Enviro Chem, the designer of the mist control system.

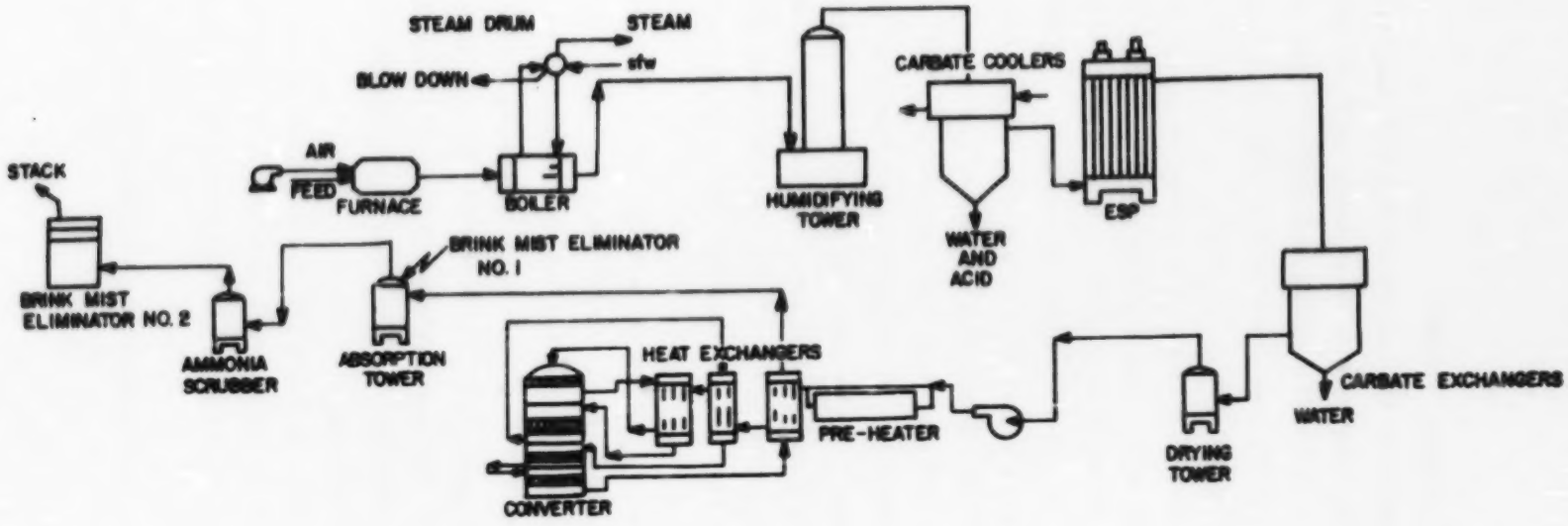

Figure 4. Gas flow sheet for sulfuric acid plant at the Collier Carbon and Chemical Corporation.

The Collier plant presents some of the same conditions--gases exposed to combustion temperatures in the presence of excess air, organic materials present in the added fuel, limited clean up of the gases prior to the cooling and humidifying tower--that existed at Bor. The dust content of the Collier gases should be lower than that of the Bor reverberatory furnace gases. While the Collier system affords less cooling of the feed gases prior to the humidifying tower, it accordingly provides a greater degree of "shock" cooling at the humidifier. Beyond the converter, however, the Collier gases are subjected to mist eliminators of high efficiency, an important factor in the control and elimination of a plume.

The ammonia scrubber and Brink mist eliminator No. 2 were recently added to the plant, primarily to control SO_2 emissions from this single absorption system. The Brink mist eliminator No. 1 was a part of the original system (36). It was designed to handle 35,000 Nm^3/hr of gas at 85°C with an efficiency of 90 percent on 0.3 to 3 micron particles, with a pressure drop of 184 mm H_2O. Installed in the top of the absorber, the mist eliminator consisted of 22 packed fiber elements. Each was a hollow core cylinder measuring 46 cm in diameter by 2.5 m high. The fibers were packed within corrosion-resistant, wire mesh enclosures. These elements were mounted vertically in the corrosion-resistant metal tube sheet installed in the absorption tower. Liquid trapped by the fibers drained from the elements at the rate it collected, permitting continuous operation.

When the plant was started in 1960, the stack met the requirements and was approved by the Los Angeles Air Pollution Control District. Visibility measurements revealed results better than the No. 2 Ringelmann chart.

Since tests of the gases leaving the mist eliminator inside the absorber showed that the gases were completely invisible, any slight opaqueness of the stack gases was attributed to mist formation in the stack or chemical reaction of the gaseous components with water vapor in the atmosphere. The unit tested 100 percent recovery efficiency on particles greater than 3 microns in diameter and 94 percent on 0.3 to 3 micron particles. Pressure drop across the unit varied between 50 and 152 mm water. The unit recovered about 770 kg of H_2SO_4 per day.

The ammonia scrubber and second mist eliminator were added to bring the plant into compliance with new SO_2 regulations. In May 1974, the Los Angeles Air Pollution Control District tested these units, sampling gases at the ammonia scrubber inlet and the Brink mist eliminator No. 2 outlet. The test results are tabulated in Table 10.

The equipment operated at steady conditions from 9:00 am until 5:00 pm on the day of the tests, May 29, 1974. Materials charged to the process were:

H_2S	12.6 metric tons per day
Alky Acid	32.6 metric tons per day
Sulfur	106.7 metric tons per day
Water	0.2 liters per second

The total rate of input materials was 685 kg/hour. The sulfuric acid production rate was 422 metric tons/day.

The mist eliminator outlet tested well under the required standards for total particulates and SO_2.

The opacity standard is less than 40 percent opacity (No. 2 on the Ringelmann Chart) except that a value as dark or darker than 40 may occur not more than three minutes per hour. Accordingly, the first reading of 40 for seven minutes is a violation. However, the haze was noted as light blue, not black as required for the Ringelmann test. The last six haze readings (Table 10) were noted as white in color. The haze thus does not quite fully conform with the color requirements for use of the Ringelmann Charts. The violation was minor, since the rating equaled 40 but did not exceed it.

The standards discussed in the previous paragraph are California standards. New Source Performance Standards, EPA Reg. 40 CFR 60.80, 60.81, 60.82, and 60.83, apply to sulfuric acid plants which produce sulfuric acid by burning sulfur, alkylation acid, etc. This plant meets two of the three Federal Standards: SO_2 emissions were less than 2 kg/ton of acid produced, and acid mist emissions were nil. The opacity standard could be considered violated. However, these standards do not apply to facilities where conversion to sulfuric acid is utilized primarily as a

Table 10

Ammonia Scrubber and Mist Eliminator
Compliance Test Results[1]

	Scrubber Inlet	Demister Outlet	Allowable Emissions		Efficiency of Controls, %
			California	Federal[3]	
Gas flowrate, Nm^3/hr, wet	31772	34660			
dry	31432	33131			
Gas velocity, m/sec	14.8	15.5			
Gas temperature, °C	60	33			
Total particulates, kg/hr	2.09	0.68			67
mg/Nm^3		19.9	147		
Solid particulates[2], kg/hr	0.50	0.68			0
SO_2, kg/hr	540	242	90		96
ppm by vol		270	2000		
kg/ton acid produced		1.37		2.0	
NH_3, kg/hr		0.05			
Acid mist, kg/hr	0.72	0.0			
kg/ton of acid produced		0.0		0.075	
% Opacity at 46-meter stack					
11:16 to 11:23 am		40	<40	<20	
11:23 to 11:24 am		30			
11:24		25			
11:31 to 11:36		30			
11:36 to 11:38		20			
11:38 to 11:41		30			
16:14 to 16:15		30			
16:15 to 16:17		20			
16:17 to 16:20		25			

1. Air Pollution Control District, County of Los Angeles, Test No. C-2127, May 29, 1974.
2. Solid particulates = Total particulates - sulfuric acid
3. New Source Performance Standards

means of preventing emissions of sulfur compounds. At a copper smelter, the appropriate standard for opacity is < 20 percent opacity, given in Standards of Performance of Copper Smelters, Federal Register, January 15, 1976 (p. 2339).

SECTION 6

LITERATURE REVIEW

General information relevant to the causes and control of emissions, and plumes, from sulfuric acid plants has been compiled from the technical literature; articles reviewed are listed in the Bibliography.

6.1 EMISSIONS FROM CONTACT SULFURIC ACID PLANTS

The Manufacturing Chemists Association and the Public Health Service conducted a study of atmospheric emissions from sulfuric acid manufacturing processes.(25) Emissions and operating data were compiled for about 12 percent of the total number of establishments. These data help to assess the general performance of an acid plant. Two tables from this study are appended to this report: Table A1, Emissions and Operating Data for Contact Sulfuric Acid Plants with Mist Eliminators; Table A2, Acid-Mist Collection in Absorber Stacks of Contact Sulfuric Acid Plants. Several types of plants are included in the tables. The data indicate that plants at that time (1965) were achieving 96.9 to 98.5 percent conversion of SO_2 to SO_3. Typically, SO_2 concentrations in the stack gases ranged from 0.14 to 0.35 percent. Plume opacity ranged from none to light. Acid mist leaving the mist eliminator ranged from 6.3 to 826 mg/m^3 (0.18 to 23.4 mg/scf). Mist leaving the absorber ranged from 35 to 1723 mg/m^3 (1 to 48.8 mg/scf) except that one plant that used no water removal facility for the discharge combustion gases showed a mist level of 89,450 mg/m^3 (2533 mg/scf). The mist particles leaving the absorber ranged from 7 to 70 percent less than 3 microns in diameter. By comparison, the Collier Plant (Section 4) showed only 20 mg/m^3 of total particulates.

Existing plants, at the time of this study, were achieving lower conversion and less emissions control than could have been attained had the plants been using the best processing techniques then available. A German double absorption contact plant offered in 1963 claimed 99.7 percent conversion at approximately the same investment as the single adsorption plants then in operation.(38) (The Bor plant uses a single absorption process.)

6.2 SULFURIC ACID MIST FORMATION

In most contact plants, dryers reduce the moisture content of gases entering the converter to 106 mg/m^3. As these gases leave the converter,

acid mist will form if the temperature falls below the dew point.(25) The mist tends to be of very fine particle size and is not absorbed in the SO_3 absorber. Theoretically, moisture in the dried gas at this level could generate 530 mg/m^3 of mist.

Traces of NO_x in the inlet gas to the absorber are reported to hinder the adsorption of SO_3.(25) NO_x is also believed to promote the formation of SO_3 prematurely in the feed gases before they enter the humidifying tower. Upon cooling, the SO_3 forms mist by reacting with the water present. NO_x is formed by combustion of fuels at high temperature with excess oxygen. Brink found that the acid mist collected on the fiber mist eliminator of a typical hydrogen sulfide-spent acid fed plant contained two percent nitrogen as nitric acid.(27) ESP's may produce NO_x if arcing occurs.

Plants which produce oleum face problems of mist formation; generally the amount of mist varies directly with the proportion of plant output in the form of oleum and the strength of the oleum produced. The extent of cooling of the converter gas before it passes to the oleum tower influences the particle size and hence the visibility of the mist in the exit gases.

The visibility of an acid mist depends more on particle size than on concentration. High proportions of mist particles less than 3 microns in diameter could cause a heavy plume if present in concentration greater than 35 mg/m^3.(28) Mists containing particles up to 10 microns in diameter have been observed, since there is visible light scattering. Conversely, if the particle size exceeds 10 microns, then the plume may not be visible at concentrations of up to 176 mg/m^3.

Unabsorbed SO_3 can be hydrated to mist when exposed to moist air. A detached plume is an indication that this is taking place.

6.3 CONTROL OF MISTS

Numerous types of entrainment separators are available to collect mists: cyclones, wire mesh impingers, ESP's, fiber mist eliminators, and venturi scrubbers. Currently designed acid plants now employ any of these that are considered appropriate. Figure 4 shows their typical collection efficiencies.

Fiber mist eliminators effect the elimination of submicron particles of mist by providing a surface with which the particles can collide under Brownian movement. A particle of 0.1 micron dimeter will have about five

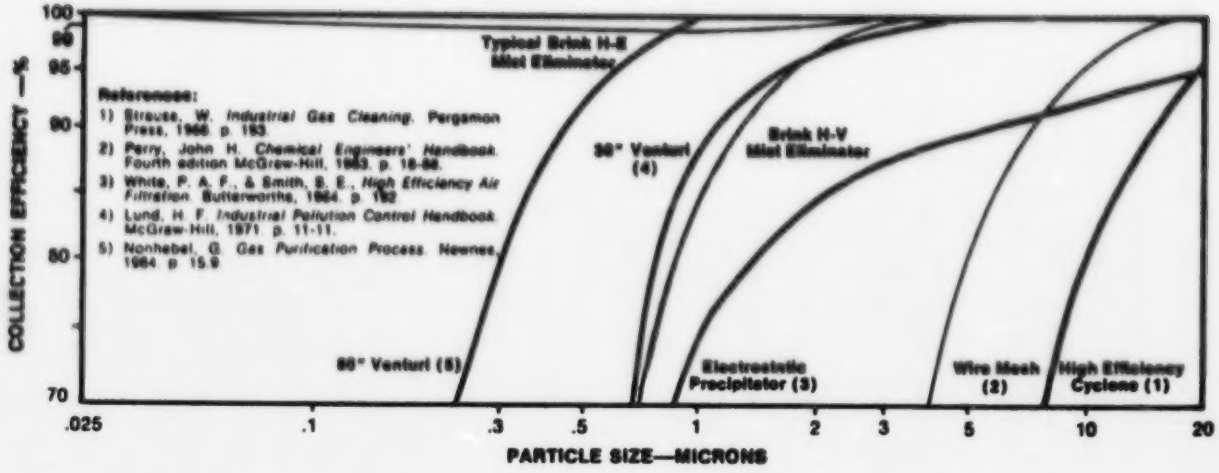

Figure 5. Typical collection efficiencies of particulate collectors.

times the Brownian displacement (caused by collision with gas molecules) of a 1 micron particle. This increases the probability that the particle will collide with a fiber and be collected. Larger particles are collected by impaction and interception. Mist particles collected on the fibers unite and form a film which moves horizontally through the fiber bed, propelled by the gases. Once on the inside surface, the liquid moves downward to a collection seal (or directly into an absorber if the eliminator is installed directly above it) and are returned to the process. (32, 34)

Mineral wool filters show improved acid mist collection efficiency if moisture is present on the surface.(31) This material is characterized by small fiber diameter (4 microns), and resistance to high temperature (540°C).

ESP's show good efficiency as mist collectors in acid plants; the wire and tube design is the most frequently used. Usually, two ESP's in series are prescribed because the first tends to overload, and when this occurs, process adjustments can be made with the second ESP as effective backup. For acid plants, tests have shown (29) particle migration velocities of 0.21 to 0.18 m/sec, with attendant efficiencies of about 99.7 percent. For known migration velocities, the probable efficiency can be calculated using the Duetch equation.(35) Stopperka(35) has investigated the effect of design and gas moisture content on the collection of acid mist with ESP's.

A relatively new method, fog filtration, is being used to remove contaminants such as H_2SO_4 mists in TNT plant streams. This is essentially a high pressure scrubber in which vaporized water cleans the gas in silo-shaped chambers spaced about 15 meters apart.(30) This gas cleaning system applies as follows: partially clean gas from ESP's is fed to one of three fog filters where it is washed under high pressure, pulled downward in a swirling motion, drawn through a pipe and expelled. The effluent water flows from the bottom of the filter to a collecting basin. The fog particles from the high pressure nozzle provide an induced draft; all gas thus entrained moves with the velocity of the fog particles. These particles wet the mist particles and carry them out of the air by simulated cyclone action.

If for any reason, NO_x in the gases cannot be reduced by control of combustion, and adequate mist elimination cannot be elected, a method for NO_x removal from the g[illegible] has been investigated (33) in which the gas is passed over coal particles which absorb the NO_x. The chief application of this method appears to be in nitric acid plants, where the absorbed NO_x contributes to higher process efficiencies.

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APPENDIX

Table A1. Emission and Operating Data for Contact Sulfuric Acid Plants with Mist Eliminators

Table A2. Acid-Mist Collection in Absorber Stacks of Contact Sulfuric Acid Plants

TABLE A1. EMISSION AND OPERATING DATA FOR CONTACT SULFURIC ACID PLANTS WITH MIST ELIMINATORS[a]

Plant type	Sulfur			Combination			
	Air dil.	No air dil.					
Raw material	Molten dark			Spent acid, H_2S, and supplemental sulfur			
Plant number	1	2A[b]	2B[b]	3A	3B	3C	4
H_2SO_4 production, tons/day	961	150	150	240	240	219	133
Percent of maximum capacity	96	68	68	100	100	91	60
Oleum made, % of output	0	0	13	0	0	0	56
Oleum made, % of free SO_3	0	0	30				40
Stack gas temperature, °F[d]	186	165	166	180	180	76	76
Stack gas rate, Mscfm	58	7.4	7.4			12	7
SO_2 entering converter, vol %	8.0	8.0	8.0			8.2	8.4
SO_2 in stack gas, vol %	0.14	0.19	0.20	0.34	0.35	0.26	0.17
Conversion of SO_2 to SO_3, %	98.5	97.6	97.5			97.2	98.2
SO_2 emitted, tons/day	10.4	1.8	1.9			4.0	1.5
Type of mist eliminator	Wire mesh			Electrical precipitator			
Acid mist leaving absorber, % less than 3 microns	70	7.5	62.0				
Acid mist leaving absorber, mg/scf		48.8	37.3	5.9	4.9	7.1	29.0
Acid mist leaving mist eliminator, mg/scf	6.5	3.6	23.4	0.33	0.38	0.18	0.31
H_2SO_4 collection efficiency, %		92.6	37.2	94.5	92.2	97.5	99.9
H_2SO_4 emitted, tons/day	0.60	0.04	0.27			0.003	0.003
SO_3 concentration leaving absorber mg/scf		2.1	1.0				
SO_3 concentration leaving mist eliminator, mg/scf		1.1					
Plume opacity	light	none	med.	light	light	light	light

[a]Sampling points:
Plant 6A and 6B: in horizontal duct between absorber top and exit stack.
All other plants: in duct or exit stack near top of absorber.
[b]Test data for Plant 2A and 2B were obtained by joint MCA-PHS field testing.

Plant type	Combination				Wet gas	Sulfur	Combination
Raw material	Spent acid, H_2S, and supplemental sulfur				H_2S sulfur	Molten dark	Spent acid, sulfur
Plant number	5A	5B	6A[c]	6B[c]	7	8	9
H_2SO_4 production, tons/day	300	300	265	300	100	429	272
Percent of maximum capacity	76	76	88	100	67		
Oleum made, % of output	0	2	0	0	0	13	0
Oleum made, % of free SO_3		25				21.5	
Stack gas temperature, °F[d]	175		160	180	130	176	150
Stack gas rate, Mscfm	17		14	21	11	28.0	19.3
SO_2 entering converter, vol %	9.0		7.2	7.4		8.0	7.4
SO_2 in stack gas, vol %	0.32		0.16	0.19		0.19	0.20
Conversion of SO_2 to SO_3, %	96.9		98.0	97.8		98.0	96.7
SO_2 emitted, tons/day	7.0		2.8	5.1		6.8	5.0
Type of mist eliminator		Glass fiber				Teflon® mesh	
Acid mist leaving absorber, % less than 3 microns					38		
Acid mist leaving absorber, mg/scf	1-4[e]	10-30[e]	20.6	32	2533[f]		
Acid mist leaving mist eliminator, mg/scf	0.5-2	7-9	0.23	1.9	2.3	1-2	1-4
H_2SO_4 collection efficiency, %	50.0	60.0	98.9	94.1	99.9		
H_2SO_4 emitted, tons/day	0.01-.05		0.005	0.06	0.04		
SO_3 concentration leaving absorber mg/scf	1-2						
SO_3 concentration leaving mist eliminator, mg/scf	1-2						
Plume opacity			none	faint	light	faint	faint

[c]Raw material included 48.4% spent acid, 32.8% H_2S, and 18.8% sulfur.
[d]Some of the temperatures are of acid entering the tower; however, inlet gas and acid temperatures are usually close together.
[e]High-velocity type glass-fiber mist eliminator designed for only medium performance.
[f]This "wet gas" unit utilized no water removal facilities for the discharge

TABLE A2. ACID-MIST COLLECTION IN ABSORBER STACKS OF CONTACT SULFURIC ACID PLANTS[a]

Raw material	Molten sulfur		Molten sulfur and spent acid	
Plant number	1	2	3	4
H_2SO_4 production, tons/day	420	422	272	302
Oleum, % of output	13	51	0	71.5
Oleum, % of free SO_3	21.5	30.8	0	20.0
Stack gas temperature, °F	176	123	150	163
Stack gas flow rate, Mscfm	28.0	25.1	19.3	19.7
SO_2 entering converter, vol %	8.0	8.8	7.4	8.0
Conversion of SO_2 to SO_3, %	98.0	97.5	97.6	97.8
SO_2 in stack gas, vol %	0.19	0.25	0.30	0.20
Ambient temperature (avg), °F	25	29	42	35
Ambient temperature (range), °F	8-37	25-36		
Wind direction	NW, W, SW	N, NW, W	S-SE	S-SW
Wind velocity (avg), mph	10.1	6.4	12	3
Stack plume opacity	faint	light	faint	medium
Stack gas velocity, ft/min	1990	1620	1080	775
H_2SO_4 concentration leaving absorber, mg/ft³[b]		2-4		10-12
H_2SO_4 concentration leaving Teflon demister, mg/ft³[b]	1-2	no demister	1-4	no demister
Acid drip from base of stack, lb/day	22	9	0	0
Strength of acid drip, % H_2SO_4	70.4	99.5		
Total acid mist from absorber collected in stack, wt %[c]	11.0	3.6	0	0

[a]All plants incorporate internal, packed bed "spray" eliminators as part of the standard absorber design.

[b]Acid mist concentrations are normal loadings, but were not obtained during acid drip measurements.

[c]The amounts of acid mist collected were averages from several tests conducted at each plant.

CONVERSION TO SI UNITS OF MEASURE

(°F + 459.67)/1.8	=	°K
M scfm x .4179474	=	N km^3/s
mph x .44704	=	m/s
ft/min x .00508	=	m/s
mg/f^3 x 35.3	=	$\mu kg/m^3$
lg/day x .45359	=	kg/day
tons/day x .907.185	=	metric tons/day